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## Coplanar Si[000, 440, 404] Three-Beam Diffraction. II. Precise Calculation

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### Abstract

The results of the measurement of the Ni  $K\alpha_2$  spectral-line peak with a double-crystal spectrometer with thick perfect silicon crystals in the dispersive arrangement (440 sym., +440<sup>3</sup> sym.) were published some time ago. A sharp anomaly was found in the measured rocking curve when the second crystal was adjusted for coplanar three-beam Bragg 440<sup>3</sup> diffraction. In this paper, the experimental curves are compared with the precise calculation based on the dynamical theory of X-ray diffraction.

### 1. Introduction

Coplanar three-beam Si[000, 440, 404] X-ray Bragg diffraction was first mentioned by Deslattes (1968) and was later examined by Graeff & Bonse (1977) from an interferometric point of view. The precise measurement of this diffraction by double-crystal diffractometry was performed by Pacherová & Bubáková (1987).

It was shown by Graeff & Bonse (1977) that coplanar three-beam [000, 440, 404] diffraction apparently changes the properties of the 440 reflection around the value of  $\lambda_m \sim \text{Ni } K\alpha_2$  (for which the condition for three-beam coplanar [000, 440, 404] diffraction can be exactly fulfilled) in comparison with the usual two-beam [000, 440] diffraction. In such a case, the method of having the double-crystal diffractometer arranged as a spectrometer, in which the sample crystal is adjusted to the three-beam diffraction, can be properly used in the experimental part of the treatment of this type of many-beam diffraction.

Pacherová & Bubáková (1987) showed the results of such measurement. In a narrow wavelength interval, a very sharp anomaly was found. A simple calculation describing the experiment was suggested and the necessary calculation performed to explain qualitatively the anomalous change. In the experiment, the sample surface was parallel to the (110) lattice planes. Thus, the 440 reflection was symmetric

cal and the 404 reflection was the extremely asymmetrical one, parallel to the surface.

In the calculation (Pacherová & Bubáková, 1987), the basis for the explanation of the anomalous change observed in the rocking curve was the size of the spectral window of the spectrometer created by the Du Mond method. It was also assumed that the whole reflected energy can be found only in the 440 reflection. This assumption is based on the experimental finding that no reflected energy was detected in the extremely asymmetrical 404 reflection.

Another simplification in the work of Pacherová & Bubáková (1987) lay in the expression of the fundamental system of equations of the dynamical theory itself: the linearization of coefficients was used in the calculation.

In spite of the simplifications described above, qualitative agreement of the experimental and calculated curves was found. The authors believed that a quantitative discrepancy results mainly from a non-ideal adjustment of the sample crystal to the coplanar [000, 440, 404] position because the anomalous behaviour of the rocking curve was very sensitive to the adjustment. Another reason for the discrepancy is the simplifications in the simulation of the experiment.

In this paper, we present the precise calculation of the coplanar three-beam diffraction [000, 440, 404] and its comparison with experiment.

## 2. Calculation of the reflection coefficient

The meanings of all symbols used in the following are given in detail by Penning & Polder (1968) and by Pacherová & Bubáková (1987). See also Fig. 1, where in a reminder is given of the definitions of the basic vectors used in the fundamental system of equations of X-ray diffraction.

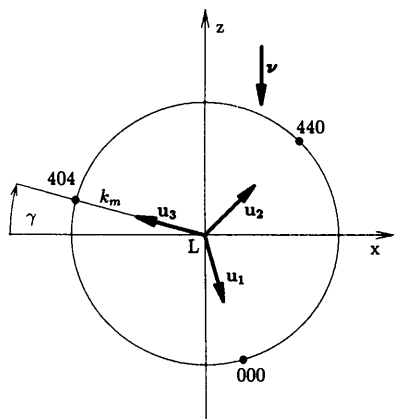


Fig. 1. Ewald's geometrical construction for coplanar three-beam [000, 440, 404] diffraction. The definition of the basic vectors  $u_1$ ,  $u_2$ ,  $u_3$  and  $\nu$ , the quantity  $k_m$  and the system of coordinates used in the calculation are shown.

The fundamental system of equations of  $n$ -beam diffraction [Penning & Polder, 1968, equation (3.8)] for the amplitudes  $Z_j$  of the waves inside the diffracting crystal can be arranged as follows for coplanar three-beam diffraction (Pacherová & Bubáková, 1987):

$$\sum_{j=1}^3 B_{ij} Z_j = 0, \quad i = 1, 2, 3,$$

which leads, for [000, 440, 404] diffraction, to the dispersion equation

$$B_{11}B_{22}B_{33} - B^2(B_{11} + B_{22} + B_{33}) + 2B^3 = 0, \quad (1)$$

in which  $(B_{ij(i \neq j)})B = C_{\sigma, \pi} \psi_{440}$  [ $C_{\sigma} = 1$ ,  $C_{\pi} = (\mathbf{u}_i \cdot \mathbf{u}_j)$ ] for all  $i, j = 1, 2, 3$  (the meaning of the coefficients  $B_{ii}$  will be treated in the following) and to the amplitude ratios  $r_2 = Z_2/Z_1$  and  $r_3 = Z_3/Z_1$ :

$$\begin{aligned} r_2 &= B(B - B_{33}) / (B_{22}B_{33} - B^2), \\ r_3 &= B(B - B_{22}) / (B_{22}B_{33} - B^2). \end{aligned} \quad (2)$$

In the experiment, [000, 440] two-beam diffraction also takes place. In the formalism of Penning & Polder (1968) used above, the dispersion equation corresponding to [000, 440] diffraction is

$$B_{11}B_{22} - B^2 = 0 \quad (1')$$

and, for the ratio of the amplitudes,

$$r_2 = -B/B_{22}. \quad (2')$$

The solution of the dispersion equation is hidden in the coefficients  $B_{ii}$  via the vector  $\Delta \equiv (-x(\theta), -z)$ , where  $\theta$  is the departure of the angle of incidence from  $60^\circ$  (the value of the Bragg angle corresponding to the wavelength  $\lambda_m = 1/k_m$ ).

Concerning the coefficients  $B_{ii}$  ( $i = 1, 2, 3$ ), we have, in practice, three possibilities for their expression. (a) They can all be expressed in the linear approximation if no extremely asymmetrical diffraction is assumed (see e.g. Bedyńska, 1973). (b) They can all be expressed in the 'circular' approximation. (c) Two of them (corresponding to not extremely asymmetrical diffraction) are expressed as in (a) and the third one (corresponding to extremely asymmetrical diffraction) is expressed as in (b). In connection with the choice of the approximation, the polynomial in variables  $x$  and  $z$  in the dispersion equation [the left side of (1)] is of degree (a) three, (b) six or (c) four. The wavefield inside the diffracting crystal is then assumed to be composed of the corresponding number of waves and, also, the boundary conditions must be set in the corresponding way.

In the linear approximation,  $B_{ii}$  can be expressed as

$$B_{ii} = B'_0 - 2(\mathbf{u}_i \cdot \Delta)/k,$$

where  $B'_0 = B_0 + 2\delta k/k$  and  $\delta k = k - k_m$  ( $B_0 \equiv \psi_0$ ).

In the 'circular' approximation,  $B_{ii}$  are expressed as (e.g. Chang, 1984)

$$B_{ii} = B_0 - (K_i^2 - k^2)/k^2,$$

where  $\mathbf{K}_i = k_m \mathbf{u}_i + \Delta$ .

Case (b) solves the problem of the [000, 440, 404] diffraction, no matter which of the beams 000, 440 or 404 is the extremely asymmetrical one. For instance, the case  $\gamma \sim -60^\circ$  corresponds to the grazing incidence beam or  $\gamma \sim 60^\circ$  to the extremely asymmetrical 440 beam. In the experiment  $\gamma \sim 0$ , i.e. the 404 beam was the extremely asymmetrical one and, because this paper is devoted to comparison of the calculation with experiment, in the present paper we consider only treatment (c) with  $\gamma \sim 0$ .

The explicit expressions for the dispersion equation (1) – equations (1a), (1b) and (1c) corresponding to cases (a), (b) and (c), respectively – are given in the Appendix.

In all subsequent calculations, the absorption of radiation in the crystals was taken into account as usual through the complex values of the coefficients  $B_0$  and  $B$ .

We consider the sample crystal to be semi-infinite. In a semi-infinite crystal, only some of the solutions  $z^j(\theta)$  of the dispersion equation (1) contribute to the wavefield inside the crystal. In the coordinate system introduced as shown in Fig. 1, these are  $z^j$  with  $\text{Im}(z^j) < 0$ . In case (c), there are two roots of the dispersion equation (1c) that satisfy this condition.

In the following boundary equations, the subscripts 1, 2 and 3 relate to the beams 000, 440 and 404, respectively. The superscripts correspond on the left side of the equations to the two roots of the dispersion equation (1c)  $z_j$  ( $j = 1, 2$ ) and on the right side to the incident and reflected beams. There are two possibilities for the beam 404:

(i) in the dispersion construction corresponding to the given wavelength  $\lambda$ , there is no intersection of the surface normal appropriate to the given angle of incidence with the vacuum 404 reflection circle, i.e. the beam 404 in the vacuum above the surface doesn't exist; or

(ii) there are two such intersections and we choose the one that corresponds to the vacuum wave vector  $\mathbf{k}_3$  directed to the outside of the crystal.

We set the boundary equations as follows:

(i)

$$\begin{aligned} Z_1^+ + Z_1^2 &= Z^i \\ r_2^1 Z_1^+ + r_2^2 Z_1^2 &= Z^{r2} \\ r_3^1 Z_1^+ + r_3^2 Z_1^2 &= 0, \end{aligned}$$

(ii)

$$\begin{aligned} Z_1^+ + Z_1^2 &= Z^i \\ r_2^1 Z_1^+ + r_2^2 Z_1^2 &= Z^{r2} \end{aligned}$$

$$r_3^1 Z_1^+ + r_3^2 Z_1^2 = Z^{r3}$$

$$\kappa_3^1 r_3^1 Z_1^+ + \kappa_3^2 r_3^2 Z_1^2 = \kappa_3 Z^{r3},$$

where the  $r_j^i$  are calculated *via* (1) and (2),  $\kappa_3^i = (\mathbf{K}_3^i \cdot \mathbf{v})$ ,  $\kappa_3 = (\mathbf{k}_3 \cdot \mathbf{v})$ . The reflection coefficient  $R$  is as usual considered to be the quantity

$$R_j = b_j |Z^{rj}|^2 / |Z^i|^2, \quad (3)$$

where  $b_j$  is the factor of asymmetry of the beam  $j$ . In the case considered,  $b_2 = b_{440} \sim 1$  and  $b_3 = b_{404} = -\kappa_3 / [k \sin(60 + \gamma + \theta)]$ .

Pacherová & Bubáková (1987) took the boundary conditions to be

$$Z_1^+ = Z^i, \quad r_2^1 Z_1^+ = Z^{r2}, \quad r_3^1 Z_1^+ = Z^{r3}$$

and, consequently, for  $\gamma = 0$ ,  $b_2 = 1$  and  $b_3 = 0$ ,

$$R_2 = b_2 |r_2^1|^2 = |r_2^1|^2, \quad R_3 = 0. \quad (4)$$

Relation (4) for  $R_2$  is valid also in the case of the not extremely asymmetrical two-beam 440 reflection, with  $r_2^1$  calculated *via* (1') and (2').

### 3. The convolution of the double-crystal rocking curve

In the ideally adjusted double-crystal spectrometer (DCS), the plane of incidence is common to all participating diffractions of both crystals. The measurement of the rocking curve involves the rotation of the sample crystal, which is in the coplanar three-beam diffraction position, with respect to the axis perpendicular to the plane of incidence.

In the DCS, the whole 'spectral window'  $\sim(\Delta\theta, \Delta\lambda)$  contributes to the detected intensity at the same time. This fact is taken into account by the convolution procedure. All the contributions affecting the character of the spectral window and the convolution procedure were described in detail by Drahoukoupil & Fingerland (1982). Let the definite mutual position of both crystals be characterized by the angle  $\beta$ . Then, the rotation is described by the quantity  $\Delta\beta$ . In the simplest form, when only the basic characteristics of the spectral window are taken into account, the convolution  $I(\Delta\beta)$  calculated in the plane of incidence only can be expressed as

$$I(\Delta\beta) = \iint_{\lambda \theta} I(\lambda) R^I(\theta, \lambda) R^{II}(-\theta - \Delta\beta, \lambda) d\theta d\lambda, \quad (5)$$

where  $I(\lambda)$  is the intensity of the spectral line. The superscripts I and II represent the first and second crystals of the DCS, respectively. If the vertical divergence  $\varphi \in (-\phi, +\phi)$  is taken into account, then

$$\begin{aligned} I(\Delta\beta) &= \iiint_{\lambda \theta \varphi} I(\lambda) R^I(\theta - \alpha(\varphi), \lambda) \\ &\times R^{II}(-\theta - \Delta\beta + \alpha(\varphi), \lambda) d\varphi d\theta d\lambda, \quad (6) \end{aligned}$$

where  $\alpha(\varphi) = 0.5\varphi^2 \tan \theta_B$  (for the symmetrical reflection,  $\theta_B$  is the Bragg angle).

#### 4. Results and discussion

We present the results of the calculation (3) for the special case  $\gamma=0$ . In Figs. 2 and 3, the values of  $R_2(\theta, \lambda)$  are illustrated with the help of contour lines. These correspond to the following values of the reflection coefficient: 0.025, 0.05, 0.1, 0.2, ..., 0.9.

It was found that  $R_3$  is not identically equal to zero as (4) suggest but, in practice, it is nonzero only in the very sharp band along the narrow branches in the  $(\theta, \lambda)$  plane of the illustration of  $R_2(\theta, \lambda)$ . In the same bands there are negligible differences in the values of  $R_2(\theta, \lambda)$  calculated according to (3) or (4).

Describing the symmetrical two-beam diffraction [000, 440], the same representation of the reflection coefficient would be composed of the parallel lines going from the bottom left corner to the upper right one (these lines are the asymptotes of the corresponding lines shown in Figs. 2 and 3).

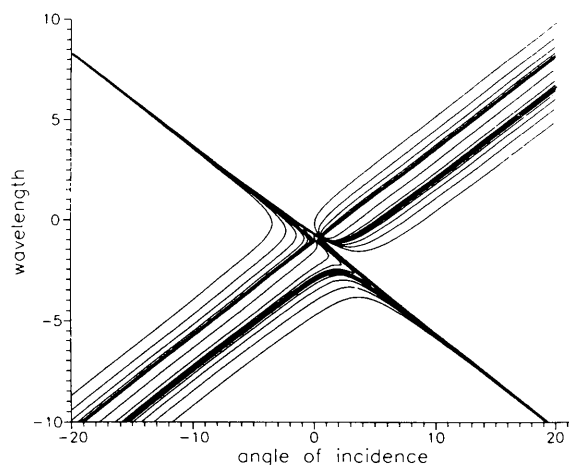


Fig. 2. The reflection coefficient of the 440  $\sigma$  symmetrical reflection corresponding to coplanar three-beam [000, 440, 404] diffraction in a silicon crystal in contour-line form. The area illustrated is  $\theta \in (-20'', +20'')$ ,  $\lambda \in (\lambda_m - 10\text{fm}, \lambda_m + 10\text{fm})$ .

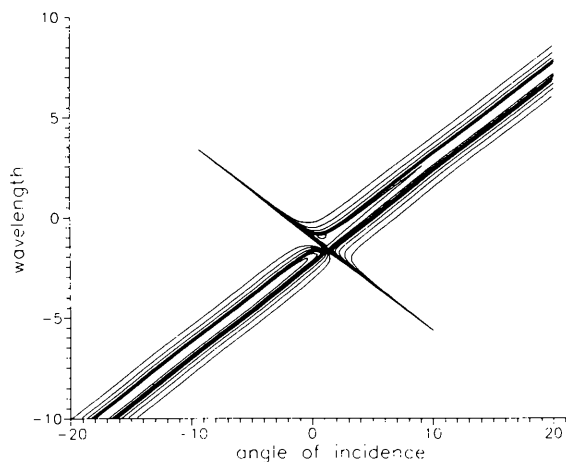


Fig. 3. The same as for Fig. 2 for  $\pi$  polarization.

In Fig. 4, the comparison of the calculated [(5)] and the experimental anomalies of the (Si 440 sym., + Si 440<sup>3</sup> sym.)/Ni  $K\alpha_2$  rocking curve is shown. The value 100% corresponds to the same rocking curve with both crystals adjusted to the two-beam diffraction.

For several points, the more precise calculation [(6)] was performed for the value of  $\phi \approx 0.5^\circ$ , which leads to the value  $\alpha(\phi) \approx 20''$ .

The agreement of the calculated and experimental curves is quite good in this more precise calculation. We can therefore conclude that the adjustment of the sample crystal in the experiment published by Pacherová & Bubáková (1987) in the coplanar three-beam [000, 440, 404] diffraction was accurate and that the anomaly obtained shows the effect of the three-beam diffraction of the sample crystal in its full extent. Calculation with the vertical divergence brings only insignificant changes to the calculated rocking curve.

On the other hand, the agreement obtained can be considered to confirm that the present calculation of the reflection coefficient for the three-beam diffraction studied is correct.

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#### APPENDIX

The coordinate system  $xz$  in the plane of incidence is introduced as usual so that axis  $z$  is parallel to the

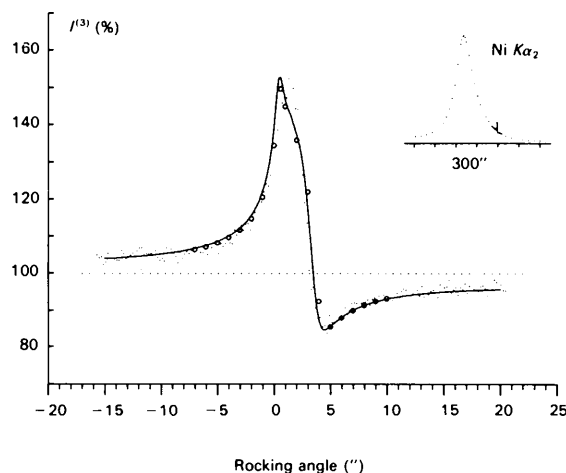


Fig. 4. Comparison of the calculated and the experimental anomalies arising on the [Si 440 sym., + Si 440<sup>3</sup> sym.]/Ni  $K\alpha_2$  rocking curve when the second crystal of the DCS is adjusted to the coplanar three-beam [000, 440, 404] diffraction position. ●●● experimental points; — calculated curve without vertical divergence; ○○○○ calculated points with vertical divergence. In the upper part, the whole Ni  $K\alpha_2$  spectral line with the anomaly schematically drawn is shown.

surface normal  $\mathbf{v}$ . With regard to the orientation of the sample crystal in the experiment we chose the coordinate set as shown in Fig. 1. We can express the dispersion equation (1) explicitly:

$$C_3^a z^3 + C_2^a z^2 + C_1^a z + C_0^a = 0, \quad (1a)$$

$$C_3^a = -2 \sin \gamma (3 - 4 \sin^2 \gamma),$$

$$C_2^a = 3[2 \cos \gamma (1 - 4 \sin^2 \gamma)x - kB'_0],$$

$$C_1^a = 6 \sin \gamma (3 - 4 \sin^2 \gamma)x^2,$$

$$C_0^a = 2 \cos \gamma (1 - 4 \sin^2 \gamma)x^3 - 3kB'_0 x^2 + k^3(B_0'^3 - 3B_0' B^2 + 2B^3);$$

$$C_6^b z^6 + C_4^b z^4 + C_3^b z^3 + C_2^b z^2 + C_1^b z + C_0^b = 0, \quad (1b)$$

$$C_6^b = -1,$$

$$C_4^b = -3[x^2 - k^2(1 + B_0)],$$

$$C_3^b = -2 \sin \gamma k_m^3 (3 - 4 \sin^2 \gamma),$$

$$C_2^b = 3[-x^4 + 2k^2(1 + B)x^2 + 2k_m^3 \cos \gamma (1 - 4 \sin^2 \gamma)x + C^b],$$

$$C_1^b = 6k_m^3 \sin \gamma (3 - 4 \sin^2 \gamma)x^2,$$

$$C_0^b = -x^6 + 3k^2(1 + B_0)x^4 - 2k_m^3 \cos \gamma (1 - 4 \sin^2 \gamma)x^3 + 3C^b x^2 + k^6[(1 + B_0)^3 - 3(1 + B_0)B^2 + 2B^3] - k_m^6 + 3k_m^2 C^b,$$

$$C^b = k^2\{k_m^2(1 + B_0) - k^2[(1 + B_0)^2 - B^2]\};$$

$$C_4^c z^4 + C_3^c z^3 + C_2^c z^2 + C_1^c z + C_0^c = 0, \quad (1c)$$

$$C_4^c = 3 - 4 \sin^2 \gamma,$$

$$C_3^c = 2 \sin \gamma [4x \cos \gamma + kB'_0 - k_m(3 - 4 \sin^2 \gamma)],$$

$$C_2^c = 2x^2 + 2 \cos \gamma [3k_m(1 - 4 \sin^2 \gamma) - kB'_0]x - [k^2(1 + B_0) - k_m^2](3 - 4 \sin^2 \gamma),$$

$$-4kk_mB'_0 \sin^2 \gamma - k^2(B_0'^2 - B^2),$$

$$C_1^c = 2 \sin \gamma (4 \cos \gamma x^3 + [kB'_0 + 3k_m(3 - 4 \sin^2 \gamma)]x^2 + 4 \cos \gamma \{kk_mB'_0 - [k^2(1 + B_0) - k_m^2]\}x - C^c),$$

$$C_0^c = -(1 - 4 \sin^2 \gamma)x^4 - 2 \cos \gamma [kB'_0 + k_m(1 - 4 \sin^2 \gamma)]x^3 + \{[k^2(1 + B_0) - k_m^2] \times (1 - 4 \sin^2 \gamma) - 4kk_mB'_0 \cos^2 \gamma - k^2(B_0'^2 - B^2)\}x^2 + 2 \cos \gamma C^c x - k^2 k_m^2 (B_0'^2 - B^2) + k^4 \{B^2 [2B - (1 + B_0)] + B_0' [B_0'(1 + B_0) - 2B^2]\},$$

$$C^c = k\{k^2[B_0'(1 + B_0) - B^2] - k_m^2 B_0' - kk_m(B_0'^2 - B^2)\}.$$

The coordinate  $x$  is related to the angle of incidence of the incident wave by

$$x(\theta) = k_m \cos(60 + \gamma) - k \cos(60 + \gamma + \theta).$$

When the incident beam is not the extremely asymmetrical one, the function  $x(\theta)$  can be expressed as the linear one

$$x(\theta) = -\delta k \cos(60 + \gamma) + k\theta \sin(60 + \gamma).$$

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## Dynamical X-ray Diffraction from Imperfect Crystals: a Solution Based on the Fokker–Planck Equation

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### Abstract

A stochastic model of crystal defects is incorporated into a Fokker–Planck equation describing dynamical

X-ray diffraction from imperfect extended-face crystals. The Fokker–Planck equation is solved by forming a set of complex moments describing the reflectance fluctuations in the crystal. This leads to